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ANNUAL REPORT

1 May 1962 to 1 May 1963

RESEARCH ON CESIUM VAPOR

Prepared for:

Office of Naval Research Department of the Navy Washington 25, D. C.

Contract No. Nonr-3805(00)

EOS Report 3130-Annual

14 June 1963

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ELECTRO-OPTICAL SYSTEMS, INC. Pasadena, California

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#### ABSTRACT

#### DOMINANT IONIZATION PROCESS IN CESIUM VAPOR THERMIONIC CONVERTER PLASMAS

The results of an investigation into the absorption of resonance radiation in cesium vapor and the effect of resonance radiation on the ionization processes in cesium vapor plasmas are reported. A model for the dominant ionization processes is proposed that appears to be applicable to the cesium pressure range and other conditions found in cesium vapor thermionic converters. It has been found that the atomic resonance absorption bandwidths at the 8521 Å and 8943 Å resonance lines are much greater than previously reported. A molecular absorption band near 7600 Å was also observed which can contribute significantly to the energy input to a cesium vapor plasma. At high cesium pressure (greater than 0.3 mm Hg) and emitter temperatures (greater than 1800°K) the absorption of resonance radiation may be the most significant energy input into a cesium vapor thermionic converter plasma leading to the formation of the cesium molecular ion as the dominant ionic species in the plasma according to the relationship:

$$Cs^* + Cs^*$$
 (heat of formation  $Cs_2$ )  $\rightarrow Cs_2^+ + e$ 

Measurements on resonance radiation decay times and cesium ionization in the presence of resonance radiation are still in progress.

#### ACKNOWLE DGMENT

The work performed under this contract has been under the technical supervision of Cmdr. J. J. Connelly, Jr. of the Office of Naval Research, Power Branch.

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# NOMENCLATURE

С	Speed of light
D <sub>i</sub>	Molecular ion dissociation energy
D <sub>m</sub>	Molecule dissociation energy
е	Charge of electron
h	Planck's constant
I abs	Intensity of absorbed radiation
Io	Incident light intensity
I,	Intensity of transmitted light through absorption cell
Ja	Atomic ionization potential
J <sub>m</sub>	Molecular ionization potential
k	Boltzmann's constant
k <sub>\lambda</sub>	Absorption coefficient as a function of wavelength
k <sub>o</sub>	Absorption coefficient as a function of frequency
L	Thickness of absorbing layer
N	Atoms excited in a volume one cm <sup>2</sup> multiplied by gap spacing in cm.
P	Pressure
P <sub>v</sub>	Power radiated by black body
Pabs	Power available for absorption
r	Internuclear separation
T	Emitter temperature
<sup>ປ</sup> ູ <sub>ບ</sub>	Energy density of radiating black body
v <sub>o</sub>	Applied voltage
v <sub>p</sub>	Positive column drop
Wrad	Energy radiated into plasma
€ <sub>T</sub>	Total emissivity
€ <sub>λ</sub>	Spectral emissivity
ΔVc	Emitter fall
<b>∆Va</b>	Collector fall
λ	Wavelength of radiation
δλ	Half height peak width of absorption lines

# NOMENCLATURE (continued)

υ	Radiation frequency
Ф	Emitter work function (cathode)
¢ a	Collector work function (anode)
т	Resonance radiation decay time

#### SUMMARY OF RESULTS

#### A. Specific Program Accomplishments

- 1. The atomic resonance absorption bandwidths of cesium vapor at 8521 Å and 8943 Å have been measured and are shown to be much greater than previously reported.
- 2. The cesium molecule absorption bands near 7600 Å are broad enough to contribute significantly to the total radiation energy input to a cesium vapor plasma at pressures higher than approximately 1 mm Hg.
- 3. A sapphire to kovar window assembly utilizing an active alloy brazing technique was perfected for use in absorption cell construction. This assembly allowed long term, high temperature, high pressure measurements to be made without window degradation.

#### B. General Conclusions

The current investigations have shown that the atomic resonance absorption bandwidths of cesium, over the pressure ranges studied, are much greater than previously thought possible. The absorption bandwidths of the 8521 Å resonance line reported in this work range from 50 to 175 angstroms over a pressure range from .06 to 8 mm Hg. This result is to be compared with previously reported absorption widths of 0.002 to 0.5 angstroms. The absorption at the 8943 Å resonance level is indicated to be only slightly weaker than the absorption at 8521 Å.

The measurements show that a cesium molecular absorption band near  $^{\circ}$  7600 Å is greater than expected on the basis of the theoretical molecular to atomic density ratio thus accounting for an additional means of introducing energy into the cesium vapor plasma. The large atomic resonance line broadening may be due to molecular formation as well as to pressure dispersive forces.

As a result of this broad atomic resonance and molecular absorption in cesium vapor, high percentage excitation of the vapor at approximately

1.45 ev will take place. Under these circumstances the formation of the cesium molecular ion as the dominant ionic species in the plasma of a cesium vapor thermionic converter is not only plausible but probable. The ionization is hypothesized to proceed according to the following mechanism:

$$Cs* + Cs* + (heat of formation of Cs_2) \rightarrow Cs_2^+ + \bar{e}$$

The importance of the ionization mechanism hypothesized herein lies in the fact that no surface ionization of any kind is required, nor is electron impact ionization required for high emitter temperatures (greater than approximately 1800°K). The ion generation in higher temperature cesium vapor thermionic converters is essentially then an energy gift which utilizes radiation from the emitter which has generally been considered a loss in the converter. The correct utilization of this phenomenon may point the way to high efficiency low temperature cesium vapor thermionic converters wherein DC to AC conversion can take place in the device itself.

This report is essentially complimented by the work supported by the Office of Naval Research, Power Branch, at R.C.A. (Nonr 3826 (00) FBM). The two investigations differ in the respect that R.C.A. observed the radiation emanating from the cesium discharge in a thermionic converter, while the E.O.S. work entails the absolute transmission measurements of resonance radiation in the plasma. In addition, the E.O.S. investigation will measure cesium ionization cross-sections with/without resonance radiation. The R.C.A. method restricts a direct determination of the resonance radiation decay times to a very limited range.

#### C. Specific Conclusions (Tentative)

 Ion generation in high temperature cesium vapor thermionic converters is probably a stepwise process brought about by the absorption of resonance radiation which is supplied by the emitter.

\*

- 2. The dominant ionic specie in high temperature cesium vapor thermionic converter plasmas is probably  $Cs_2^+$  not  $Cs^+$ .
- 3. The resonance absorption measurements and the excitation calculations point out the shift from resonance excitation and subsequent ionization to electron impact excitation and ionization as thermionic converter emitter temperatures are lowered from approximately 2000°K to 1600°K.
- 4. With proper care, sapphire to metal window assemblies can be made which are useful in cesium vapor up to temperatures of approximately 900°K and pressures of at least 10 mm Hg. The ultra violet transmission of brazed sapphire-metal window assemblies can be greatly effected by the brazing process. The general effect is a gross loss in transmission for almost indiscernible coatings of the braze material on the window. The loss of ultraviolet transmission due to the brazing process may be corrected for by proper post braze cleaning procedures.

#### 1. INTRODUCTION

#### 1.1 General Discussion

No detailed information exists in the literature pertaining to the exact ionization and space charge neutralization processes which are dominant in cesium vapor thermionic converters. One school of thought ascribes space charge neutralization in cesium vapor converters to ions formed by surface ionization processes at the emitter (or collector) surface which then drift out into the interelectrode region and there become effective in neutralizing space charge. This particular concept is not in agreement with findings in the alkali vapor discharge literature. An extended study on low voltage rubidium vapor diode discharges (Ref. 1) reports an experiment wherein surface ionization at the collector was utilized to produce ions for injection into the interelectrode space to neutralize space charge, thus lowering the arc drop to approximately zero. The experimental findings were essentially negative in that only a gain of approximately two in space charge limited current from the emitter could be observed experimentally. This was in agreement with the predictions of Langmuir in a paper on ion and electron sheaths in gas discharges (Ref. 2). It might be observed that the potential distribution in the experimental rubidium vapor diode favored extraction of ions from the surface whereas the same situation does not hold in the case of ions generated at the surface of the emitter in a cesium vapor thermionic converter. In fact, the field is probably retarding for ions at the emitter surface in a practical converter. (see Fig. 1.1).

Another school of thought maintains that in essence the cesium vapor thermionic converter in its interesting range of operation is little more than a gas discharge diode or plasma diode with high work function emitter and low work function collector. The gas discharge nature of the cesium vapor thermionic converter was demonstrated in 1960 (Ref. 3) by

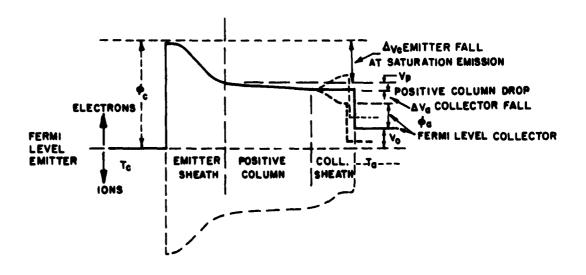


FIG. 1.1 ARC MODE VAPOR CONVERTER ENERGY LEVEL DIAGRAM NEAR MAXIMUM POWER POINT

means of breakdown measurements on an operating converter in the low emitter temperature region (1200°C). More detailed breakdown curves were presented in (Ref. 4) in the spring of 1961 at the thermionic converter contractors meeting at Cambridge, Massachusetts. Other investigators have since corroborated these results. The significant difference in interpretation deals with the nature of production of ions in the interelectrode space. It has long been known that volume ionization in a gas discharge diode occurs when electrons are accelerated across an emitter sheath gaining the kinetic energy necessary to accomplish impact ionization in the plasma many mean free paths from the sheath edge. In the case of noble gas hot cathode discharges this "emitter fall"  $\Delta V_{\rm C}$  is of the order of the first ionization potential, resonance level, or metastable level of the gas.

Measurements made (Ref. 4) on operating vapor thermionic converters give values of the emitter sheath voltage (or cathode fall) on the order of 0.6 volts to 0.8 volts in the low emitter temperature region. Measurements of arc drop in rubidium vapor hot cathode discharges (Ref. 1) give values of the tube drop less than approximately 2 volts. Similar measurements in hot cathode discharge in cesium vapor (Ref. 5) also show tube drops of less than 2.0 volts. Probe measurements on the devices of References (1) and (5) give electron temperatures in the neighborhood of  $4000^{\circ}$ K, (characteristic of cathode falls of approximately 0.5 volts) with no major potential maxima in the interelectrode space.

These measurements can be interpreted to mean that ionization is taking place by stepwise processes involving the capture of resonance radiation. This latter process almost certainly involves one (or two) cesium resonance levels near 1.45 ev (see Figure 1.2). One such stepwise ionization process may be represented by the following equation:

$$Cs^* + Cs^* + heat of formation of  $Cs_2 \rightarrow Cs_2^+ + \bar{e}$   
(1.45 ev) + (1.45 ev) + .45 ev  $\rightarrow$  3.28 ev + internal energy$$

#### ENERGY LEVEL DIAGRAM OF CESIUM

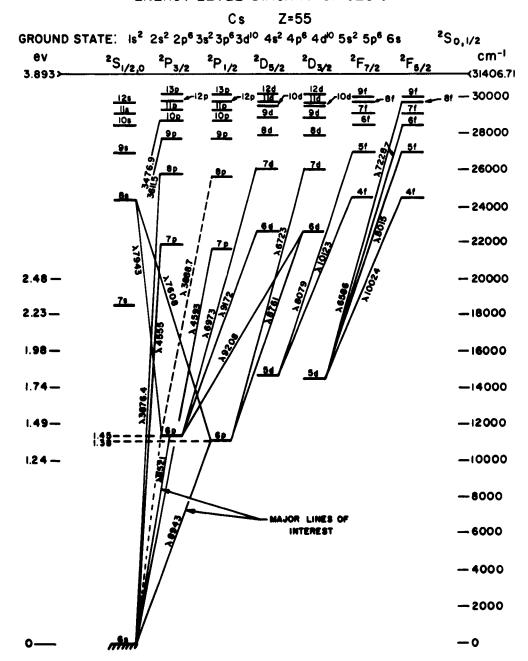


FIG. 1.2 CESIUM 1

The formation of the cesium molecular ion at an energy of roughly 3.28 ev has been given in the literature. This would require an emitter (cathode) fall on the order of only a few tenths of an electron volt, necessary only to draw saturation electron emission across the emitter sheath, if the cesium vapor existed in a state of high percentage excitation at the resonance level. In the case of low temperature emitters only a small percentage of the energy radiated from the emitter exists in the 0.85 micron band and the excitation energy would have to come from electron impact (and a resultant emitter fall of up to approximately 1.4 volts). As an emitter temperature increases, the peak of the radiation from the emitter shifts to shorter wavelengths according to the Wein displacement law  $(\lambda_m = \frac{a}{T})$  and substantially more energy is available near the cesium resonance wavelength (8500 Å). In a limiting case at high emitter temperature (2000°K) one might expect nearly all the excitation and ionization in a cesium vapor thermionic converter to be due to radiation from the emitter.

The state-of-the-art in nuclear heat source for thermionic converters would seem to indicate that the most realistic approach (at this time) to a nuclear thermionic generator system would center around a low temperature vapor thermionic converter and a suitable reactor. Due to the fact that the emitter operating temperature (1400°K to 1600°K) for a "low temperature" vapor thermionic converter is near the maximum usable temperatures achieved in state-of-the-art nuclear reactors it is essential to understand the fundamental phenomena associated with the ionization processes in vapor thermionic converters in order to maximize converter output and efficiency in the low emitter temperature region.

The possible roles of resonance radiation and the emitter sheath voltage in the ionization processes pertaining to cesium vapor thermionic converter plasmas have been briefly outlined. Implicit in any ionization process involving photon absorption and/or electron impact is an optimum electrode spacing for a given atom density in the interelectrode space.

One would expect, and indeed finds, an optimum spacing-cesium pressure relationship in practical cesium vapor converters. For given converter geometry (planar), electrode material (molybdenum), electrode temperature, and cesium pressure, the spacing effect is clearly shown in Figure 1.3 (Ref. 4).

It is thus apparent that too large a collector to emitter spacing probably introduces plasma resistivity losses, and too small a spacing leads to inefficient impact ionization and/or resonance absorption in the emitter collector gap. The output power of a practical converter appears to be significantly reduced when the spacing is not optimum. An exact, or more exact, knowledge of the ionization processes pertinent to cesium vapor plasmas should aid in minimizing such losses and possibly give a clue to how ionization could be achieved more efficiently.

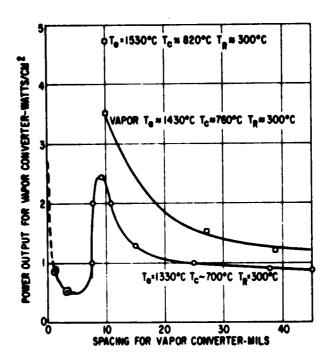


FIG. 1.3 VARIATION IN OUTPUT POWER DENSITY AS A FUNCTION OF ELECTRODE SPACING FOR SEALED-OFF VAPOR THERMIONIC CONVERTER

#### 2. BACKGROUND INVESTIGATIONS

In the first early investigations of gas discharges, it was found (Ref. 6) that ignition and maintenance potentials of low voltage arcs in alkali vapors were significantly lower than the ionization potential of the particular vapor used. It was also determined that the ignition and maintenance potentials decreased for increasing filament temperatures. The significance of these observations were not realized by investigators in the field at that time but in light of the present studies they correlate very well with our results. About this same time (1925) Foote and Mohler (Ref. 7) of the National Bureau of Standards, were studying photoionization of cesium vapors. The spectrum range of interest was 2500 - 3500 A. In this investigation Mohler observed that there was not a sharp cut off of the ionization current at the series limit of 3184 % corresponding to the ionization potential of the cesium atom at 3.89 ev. Instead, Mohler was still able to detect ionization current up to 3400 Å. He interpreted this as indicating the presence of some kind of excitation and stepwise process being involved in the ionization mechanism beyond the series limit at 3184 Å.

In 1929 Franck (Ref. 8) published several theories pertaining to complete mechanisms for the ionization of cesium vapor. One of these theories involved the possibility of an excited atom colliding with a neutral atom to form the molecular cesium ion. In 1930 Mohler (Ref. 9) attempted to experimentally verify this mechanism in cesium vapor using radiation in a spectrum range from 2800 - 3800 Å. Mohler was not able to completely verify Franck's theory for an ionization mechanism consisting of the following:

$$Cs^* + Cs \longrightarrow Cs_2^+ + electron$$

Mohler, in these particular experiments, was able to eliminate several alternate mechanisms for ionization. He eliminated the possibility of

interactions between two excited atoms due to the experimentally observed linear relation between ion current and photon intensity at a constant vapor pressure for this range of wavelength. He eliminated the possibility of ionization due to inelastic collison between electrons and atoms since he found that the ion current was independent of electron current. No direct ionization due to atom thermal velocity occurred since there was no observed cell temperature effect on ion current. Mohler observed a complex pressure dependence for the ion current at constant photon intensity indicating the possibility of step-wise ionization processes.

In 1931 Freudenberg (Ref. 10) also attempted to verify Francks's theory of the ionization mechanisms in cesium vapor. He reconfirmed the absence of inelastic electron-atom collisions since he found that the ion current was independent of electron current over a range of applied voltage from .02 V to .40 V with constant cesium vapor pressure. Freudenberg showed that except for the direct atomic photoionization at wavelengths less than or equal to 3184 % (3.89 e.v.) the reaction product is not an atomic ion. Beyond the series limit at 3184 A the additional energy necessary for ionization would have to be kinetic energy obtained by thermal effects, and no temperature dependence was found. Using monochromatic light, Freudenberg studied the temperature dependence of the ion current as a function of light wavelength. He observed no increase in ionization with temperature until he reached a wavelength of 4555 X, corresponding to an energy of 2.72 e.v. At this point he observed an onset of ionization current when the ambient temperature in the cell reached 450°C, this cell temperature gives the atoms a kinetic energy equal to 0.125 e.v.

To explain the results Freudenberg developed the Born-Haber type cycle shown in Figure 2.1. Since there is no change in enthalpy in the complete cycle:

$$J_{m} + D_{i} - J_{a} - D_{m} = 0$$

where  $\mathbf{J}_{\mathrm{m}}$  is the molecular ionization potential,  $\mathbf{D}_{\mathrm{i}}$  is the molecular ion

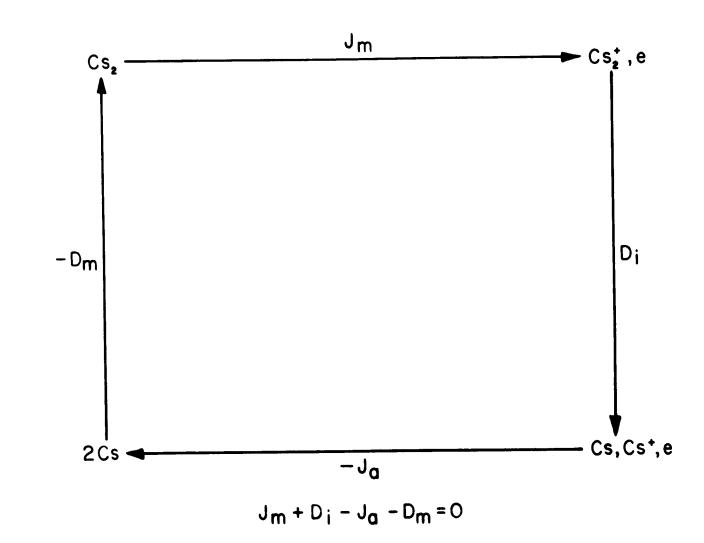


FIG. 2.1 BORN-HABER CYCLE

dissociation energy,  $J_a$  is the atomic ionization potential, and  $D_m$  is the molecule dissociation energy. From what he had observed experimentally, Freudenberg was able to develop an empirical equation (Equation 1, Figure 2-2). Equation 2 (Figure 2-2) follows directly. In Equation 2, the value 2.825 e.v. should be considered a heat of formation for the indicated reaction, completely analogous to Equations 3 and 4 which are the definitions of the dissociation and ionization energies of the molecule. This immediately relates the dissociation energy to the ionization energy through Equation 5. At the time this work was performed the dissociation energy of the cesium molecule was not known exactly. On the basis of available information on the other alkali vapors Freudenberg estimated the molecular cesium dissociation energy to be in the range from .3 to .4 e.v. On this basis the ionization potential of the molecule thus lies between the limits 3.1 e.v.  $< J_m < 3.2$  e.v. He further contended that if Equation 2 were correct, it would take higher thermal environments to observe an ion current as the incident radiation increased in wavelength. Also by increasing the wavelength of the incident light to 8521 X, which corresponds to the resonance level of cesium at 1.45 e.v., two colliding excited atoms should have sufficient energy to create the molecular ion. This, Freudenberg was able to observe experimentally. Furthermore, he was able to show that under resonance radiation the ion current increased as the square of the photon intensity at constant cesium vapor pressure. This indicated a two body collision process whereas there was only a linear relation for the lower wavelengths.

In 1934, Loomis (Ref. 11) spectrographically determined the molecular dissociation energy ( $D_{\rm m}$ ) as .45 e.v. and using Freudenberg's equations calculated a molecular ionization potential of  $J_{\rm m}=3.28$  e.v. Finkelenburg (Ref. 12) in 1938 confirmed Loomis' dissociation energy determination of 0.45 e.v. and Pauling (Ref. 13) in 1934 indicated 0.46 e.v. as the dissociation energy. Figure 2.3 summarizes this earlier work.

$$Cs^*(\lambda = 4555 \text{ Å}) + Cs + 0.125 \text{ ev} = Cs_2^+ + \text{e}$$
 (1)

$$Cs + Cs + 2.825 = Cs_2^+ + e$$
 (2)

$$Cs + Cs = Cs_2 + D_m$$
 (3)

$$Cs_2 + J_m = Cs_2^+ + e$$
 (4)

$$D_{m} - J_{m} + 2.825 = 0 (5)$$

FIG. 2.2 DETERMINATION OF DISSOCIATION AND IONIZATION ENERGIES OF THE CESIUM MOLECULE

INVESTIGATOR	CESIUM PRESSURE	MECHANISMS	RESULTS	METHODS
MOHLER (1930) (REF. 7,9)	.00025-,14mm	Cs*+Cs - Cs <sub>2</sub> +e λ=3100 TO 3900 Å	D <sub>m</sub> = 0.26 ev D <sub>i</sub> = 0.7 ev J <sub>m</sub> = 3.43 ev	SPACE CHARGE NEUTRALIZATION
FREUDENBERG (1931) (REF. 10)	.07535mm	$Cs^{*}+Cs^{*}-Cs^{+}_{2}+e$ $\lambda = 8521 \text{ Å}$ $Cs^{*}+Cs-Cs^{+}_{2}+e$ $\lambda = 4555 \text{ Å}$	D <sub>m</sub> = 0.35 ev D <sub>i</sub> = 1.05 ev J <sub>m</sub> = 3.17 ev	SPACE CHARGE NEUTRALIZATION
LOOMIS & KUSCH (1934) (REF. 11)	.03 - 2 m m		D <sub>m</sub> = 0.45 ev J <sub>m</sub> = 3.28 ev	SPECTROGRAPH
FINKELENBURG & HAHN (1938) (REF. 12)			D <sub>m</sub> = 0.45 ev	SPECTROGRAPH
PAULING (1934) (REF. 13)			D <sub>m</sub> = 0.46 ev	CALCULATION

FIG. 2.3 SUMMARY OF CESIUM INVESTIGATIONS

#### 3. SPECIFIC PROGRAM

The primary objectives in the current studies of the absorption spectrum of cesium are: (1) a determination of the amount of energy which can be absorbed by cesium vapor from the emission spectrum characteristic of the radiation from thermionic emitters. (In. particular, it is desired to know the absorption properties of cesium vapor in the spectral region between  $7000 - 10,000 \, \text{Å}$  as a function of vapor pressure and container dimensions), and (2) a knowledge of the ionization excitation mechanisms, both atomic and molecular, which apply to the vapor as a consequence of absorption of radiation.

#### 3.1 Resonance Absorption in Cesium Vapor

#### 3.1.1 Theory

The amount of radiant energy that a gas, occupying a known volume at a known vapor pressure, can absorb may be computed if the absorption coefficient of the gas as a function of frequency is known and if the spectral distribution of radiation incident on the sample is given. The observed profile or shape of an absorption line of a monatomic gas is strongly influenced by a number of factors including, for example, temperature, the frequency of collision between the absorbing atom and atoms of the same or different kind, the frequency of collision between the absorbing atom and positively charged ions or electrons, natural uncertainty in the energy levels involved in the optical transition and instrumental broadening of the absorption line.

If a continuum of radiation is passed through an absorption cell containing a monatomic gas at a given vapor pressure, an attenuation in the intensity of the beam will be observed in the region of a resonance line of the atom. The intensity of the transmitted radiation will exhibit a frequency distribution similar to that depicted

in Figure 3.1. The absorption coefficient of the gas (k,) is defined by the relation:

$$I_{v} = I_{o} e^{-k}v^{L}$$
 (3.1.1)

where L is the thickness of the absorbing layer.

If L is measured in cm, then k, has the dimensions of cm<sup>-1</sup>. Using Equation 3.1.1 in conjunction with the measured transmitted intensity, k, may be determined as a function of frequency and the relationship can be plotted graphically as is shown in Figure 3.2.

An experimental determination of the absorption coefficient frequency contour is of fundamental importance not only because it serves as a measure of the amount of radiation that the sample absorbs, but also by virtue of the fact that the half-width,  $\Delta v$ , is inversely proportional to the lifetime of the individual excited state. In addition, the qualitative shape of the contour gives an indication of the relative importance of the various perturbations that the absorbing atoms are subjected to.

The most interesting, but complicated, form of line broadening is that caused by collisons between the absorbing atom and other neutral or electrically charged particles. A great deal of experimental and theoretical work has been entered into in recent years in an effort to understand this phenomenon. The difficulty of explaining all these effects can be appreciated by observing the change in the shape of the absorption line with pressure and the appearance of extraneous bands, as illustrated in Figure 3.3.

While it is customary to reduce an experimentally determined absorption coefficient  $(k_{\lambda})$  to standard conditions (Ref. 14), such a reduction is not always valid and is essentially meaningless for pressure-broadened absorption lines of the type which will be the subject of this discussion.

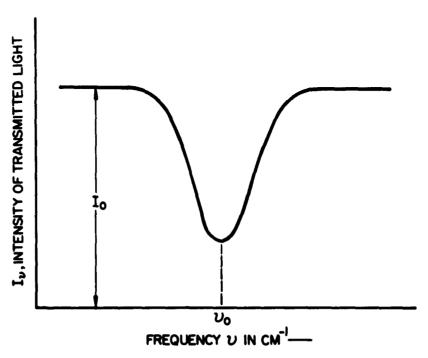


FIG. 3.1 GENERALIZED ABSORPTION LINE SHOWING INTENSITY OF TRANSMITTED LIGHT AS A FUNCTION OF WAVE-NUMBER (FREQUENCY)

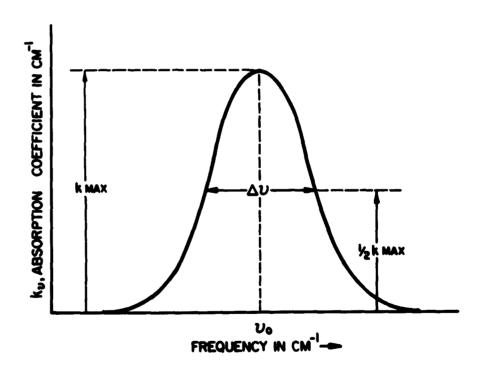


FIG. 3.2 GENERALIZED ABSORPTION COEFFICIENT-FREQUENCY CONTOUR

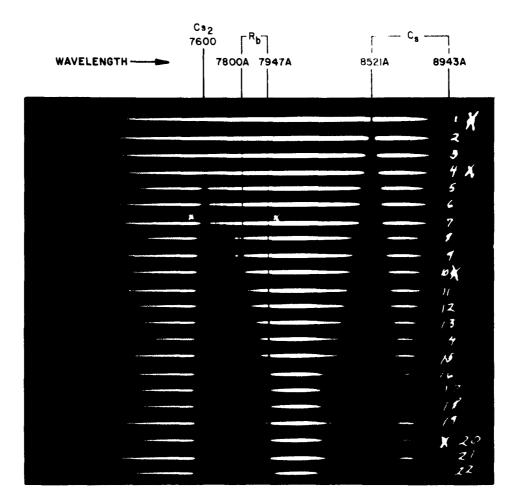


FIG. 3.3a ABSORPTION SPECTRA OF CESIUM VAPOR INDICATING THE EFFECT OF VAPOR PRESSURE ON LINE WIDTH OVER A CESIUM VAPOR PRESSURE RANGE FROM  $4\times10^{-2}$  TO 8 mm Hg.

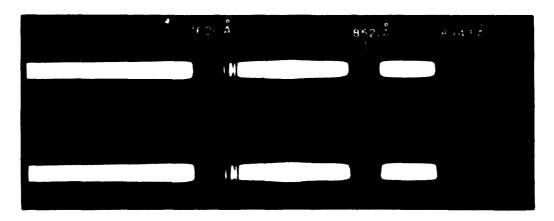


FIG. 3.3b MOLECULAR AND ATOMIC ABSORPTION SPECTRA INDICATING RELATIVE LINE WIDTH BETWEEN THE TWO ATOMIC RESONANCE LINES AT 8521 and 8943 Å

Once  $k_{\lambda}$  has been determined for a given path length, L, the rate at which the energy lying between  $\lambda$  and  $\lambda+d\lambda$  is removed from a light beam by a sample of dimensions (1 x L)cm<sup>3</sup> can be computed from:

$$I_{abs (\lambda)} = I_o(\lambda) \left[ 1 - e^{-k} \lambda^{L} \right]$$
 (3.1.2)

providing that the value of  $k_{\lambda}$  used in equation (3.1.2) was obtained experimentally by measuring the transmittance of a gas sample which was under identical conditions of temperature, pressure, and concentration of foreign gases as the gas for which the calculation is made. This restriction is invoked for pressure-broadened lines but is not generally necessary for atomic absorption lines. For example, within the temperature and pressure range where the influence of interatomic forces are neglibile, the same  $k_{\lambda}$  may be applied to gas samples under a variety of conditions of temperature pressure, and contamination.

Generally speaking, at total vapor pressures of less than latm, atomic absorption lines can be expected to be characteristically narrow and the wavelength interval over which energy may be removed from a beam of radiation passing through an atomic gas is usually of the order of a few angstrom units or less, for any given atomic transition.

The resonance absorption lines (that is, the first number of the principal series of absorption lines) of the alkali metals present an exception to the generalization stated above since relatively long-range interatomic perturbations come into play which result in significant variations in the quantized energy levels of the absorbing atoms. As a result, the resonance absorption lines are broadened to an unusually large extent. The manner in which the interatomic forces give rise to line broadening can be illustrated with the help of Figure 3.4, in which the potential energy of a system composed of two cesium atoms is plotted against the interatomic separation.

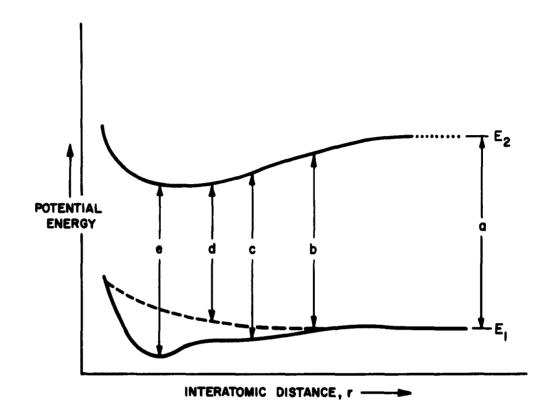


FIG. 3.4 SCHEMATIC TRENDS OF POTENTIAL ENERGY CURVES DUE TO INTERATOMIC FORCES

When a cesium atom is not subject to the perturbing fields due to the neighboring atom, the energy difference  $\mathbf{E}_2$ - $\mathbf{E}_1$  determines the wavelength of the radiation which is absorbed by the atom. This is the case if the two atoms are separated by an infinite distance. At this infinite separation the width of the absorption line is determined by natural uncertainty and by the Doppler effect. The broadening due to these two effects results in a increase in line width of the order of a hundredth of an angstrom, and both of these broadening processes become negligible compared to other broadening processes which occur at distances where the interatomic forces become important.

The forces which occur between two neutral atoms of the same species which are in close proximity (i.e., at a separation of less than 300 Å) are: (1) resonance forces, (2) dispersion forces, and (3) exchange forces. The resonance and dispersion forces occur at distances where the charge distribution of the two atoms do not overlap. The exchange forces result from an overlapping of two charge distributions, and give rise to the familiar valence or binding forces which result in a stable diatomic molecule. It is important to emphasize, however, that exchange forces may also be repulsive and, in fact, for two ground-state cesium atoms the probability that a collison gives rise to repulsive forces is approximately three times as great as the probability for attractive forces.

Expressions for resonance and dispersion forces can be obtained by expanding the classical expression for the non-overlapping charge distributions and computing the quantum mechanical interaction by means of perturbation theory (Ref. 14). The resonance forces are due fundamentally to the fact that the perturbing atom is essentially indistinguishable from the absorbing atom and dominates the dispersion forces when the probability of an absorbing atom colliding with an atom of its own kind is very much greater than the probability for a collision with a foreign atom. The dominance of the resonance forces over the dispersion forces is due to the fact that the resonance forces give rise to a first-order perturbation which can be shown to be proportional to  $r^{-3}$  where r is the internuclear separation. The dispersion forces arise from a

second order effect and are proportional to r<sup>-6</sup>. Both the dispersion and resonance forces are especially large for the resonance lines of alkali atoms as can be seen by examining the main features in the expressions for the interaction energies (Ref. 14 and 15).

In addition to the r<sup>-3</sup> dependence, the principal features of the resonance forces are that: (1) their strength is proportional to a dimensionless quantity, f, which is known as the oscillator strength of the atomic transition, and (2) their sum over all magnetic quantum numbers of the two states involved in the transition, vanishes in the mean (Ref. 15). (1) explains why the cesium resonance lines are more apt to be broadened than any other absorption lines such as, for example, the mercury 2537 Å line, since the oscillator strength for the combined cesium resonance lines is about 0.98 while the oscillator strength for the mercury 2537 A line is about 0.03. It also explains why the broadening of the higher members of the cesium absorption lines, due to resonance, decreases rapidly in importance since the f number for these higher members is very small with respect to unity. (2) merely means that a resonance interaction may cause the atomic absorption line to broaden symmetrically since an increase in the transition energy is just as likely to result as a decrease in transition energy.

The dispersion forces are abnormally large for the alkali atoms (Ref. 14) since the expression for the second order perturbation energy (Ref. 16) involves a sum of terms, each of which has a denominator which depends on the difference in energy between the ground level and the excited atomic levels. These differences are relatively small for the alkali atoms and accordingly a large number of terms contribute significant amounts to the dispersion energy. Margenau (Ref. 14) has shown that for sodium and potassium the dispersion forces are important even at relatively large interatomic distances. The dispersion forces for cesium can be expected to be even stronger than for sodium and potassium since the separations between the pertinent atomic energy levels are smaller for the cesium atom than for the latter two.

In Figure 3.4, line  $\underline{a}$ , the energy difference  $E_2$ - $E_1$  refers to the sharply quantized transition that occurs when the cesium atom is not influenced by a neighboring atom. The lines connecting the energy level curves may be various lengths depending on the distance of approach of a perturbing atom at the instant of the electronic transition. Line  $\underline{b}$  represents the modification in transition energy that results at distances where the resonance forces come into play; line c represents the transition energy when resonance and dispersion forces are of the same order of magnitude. Transitions may also occur from any point on the repulsive energy curve (dashed line). For example; line d which refers to a transition from the repulsive state to the portion of the upper curve which corresponds to a region in which attractive dispersion and resonance forces are important. The net effect of these various attractive and repulsive forces on the lower and upper states of the atomic transition is a severe broadening of the cesium absorption lines at 8521 A and 8943 Å (see Figures 3.3 and 3.5, also Ref. 17).

The line e represents a transition from the stable ground state of the cesium molecule to a stable upper state. It should be noted here that the upper potential energy curve in Figure 3.4, has been greatly simplified. There are a total of eight possible curves arising from a collision between an excited cesium atom and an atom in the ground state. Several molecular transitions which correspond to the 8521 Å and 8943 Å atomic transitions may result. Herzberg (Ref. 17) has assigned the strong 7600 Å and 9000 Å molecular absorption bands to two of these transitions.

The molecular transitions can take on an important significance since they provide an additional means to introducing energy into the vapor at higher pressures.

An exhaustive search of the literature indicates that Gregory (Ref. 18) is the only previous study of the broadening of resonance absorption lines by cesium vapor. Gregory (Ref. 18) omits several important experimental details but by referring to prior work it is evident that a relatively thin absorption cell was used (a path length between windows of possibly 0.2 cm as compared to 2.5 cm used in the current set of experiments).

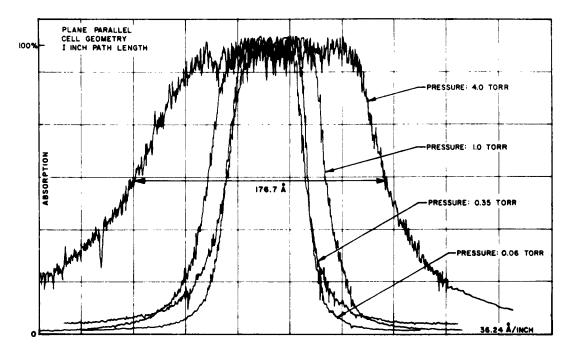


FIG. 3.5 ATOMIC RESONANCE ABSORPTION AT 8521 Å AS A FUNCTION OF PRESSURE

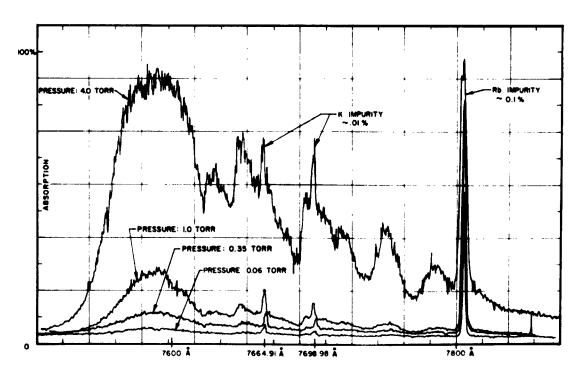


FIG. 3.6 CESIUM MOLECULAR ABSORPTION AS A FUNCTION OF PRESSURE

The data given by Gregory essentially consists of a tabulation of the half-widths of the resonance lines as a function of pressure. He does not present any information regarding absorption coefficients nor does he show any reproductions of the cesium spectra. It is difficult to make any comparison between Gregory's results and the results obtained in the current studies until the two sets of data can be reduced to a common basis.

In the current studies it was observed that for pressures greater than about 1 mm Hg the contour of the atomic resonance absorption lines become asymmetrically broadened as the perturbations due to collisions become important. As the pressure increases above a few mm Hg the atomic resonance lines become overlapped by diffuse band-like absorption features which are undoubtedly due to the Cs<sub>2</sub> molecule. The net effect of the line broadening and molecular absorption is to cause absorption in the 8521 and 8943 Å region to take place over a fairly broad wavelength region. This is readily evident in the reproductions shown in Figure 3.5. Gregory observed the diffuse features and comments that they prevent an analysis of the resonance lines in terms of purely atomic transitions.

In addition to the diffuse absorption in the vicinity of the atomic resonance lines,  $Cs_2$  gives rise to many other absorption features (Ref. 19). The most important of these are a band system in the vicinity of 7600 % (Figure 3.6) and a number of diffuse absorption features on the long wavelength side of the 8943 % line. The latter have not yet been photographed in the current set of experiments. The band system at 7600 % shows up clearly in the spectra of cesium vapor at pressures greater than 0.5 mm Hg. Several weak absorption bands have also been detected at other wavelengths but these are very weak in comparison with the 7600 % bands and will not be considered at present.

It is worthy to note that all of the molecular absorption features of  $\operatorname{Cs}_2$  reported by the workers listed in Refs. 11, 12, and 19 have been observed in the spectra obtained in the course of the present research, with the exception of the bands in the 10,000 Å region.

## 3.1.2 Experimental Method for Determination of Resonance Absorption

The absorption cell with which all the absorption measurements were made is shown in Fig. 3.7. Figure 3.8 gives an excellent indication of the transparency of the completed absorption cell. A schematic of the experimental setup for absorption measurements is shown in Fig. 3.9.

The lamp used as a radiation source was calibrated for black body temperatures of the tungsten filament at various filament currents (Fig. 3.10). This enables a closer correlation of radiation from the light source to actual operating temperatures of emitters of thermionic converters. The temperature of the lamp filament was measured over its length and was found to be completely uniform.

# 3.1.3 Experimental Results - Absorption of Resonance Radiation

An experiment was conducted using a Jarrel-Ash F/6.3 grating spectrograph to determine the effect of lamp filament temperature on resonance absorption bandwidth. It was found that there was no measureable effect on bandwidth over a filament temperature range of 1800 - 2250 K at constant cesium pressure. This indicates that the observed absorption broadening (Fig. 3.3) is completely a pressure effect over the pressure range.06 to 8 mm Hg and is not dependent on radiation intensity within this filament temperature range.

Figures 3.5 and 3.6 are densitometer recordings taken of spectra obtained with constant lamp filament temperature and cell environment temperature with the cesium pressure varied from .06 mm Hg to 4 mm Hg. Figure 3.5 shows the broadening of the 8521 Å absorption band. The 8943 Å band was not capable of being accurately measured, due to a rapid drop in plate sensitivity starting at the center of the 8943 Å absorption band. A visual comparison of the photographic plate indicated though that the 8943 Å bandwidth was only slightly less than that at 8521 Å. The half height bandwidth of the 8521 Å band at the highest pressure in Fig. 3.5 is approximately 175 Å. Figure 3.6 shows the absorption growth of the molecular band over the same pressure range.

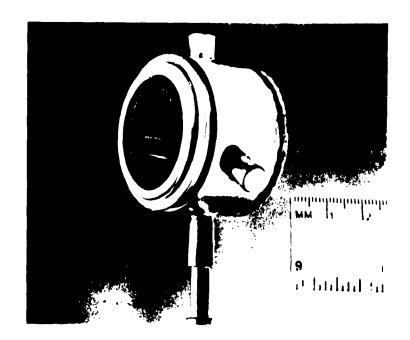


FIG. 3.7 CESIUM ABSORPTION CELL

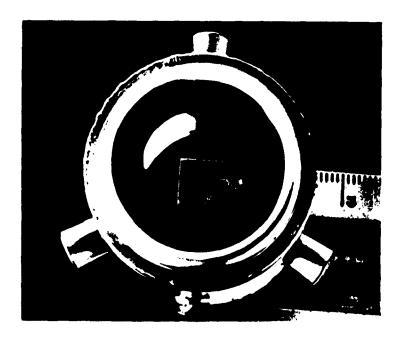


FIG. 3.8 CESIUM ABSORPTION CELL

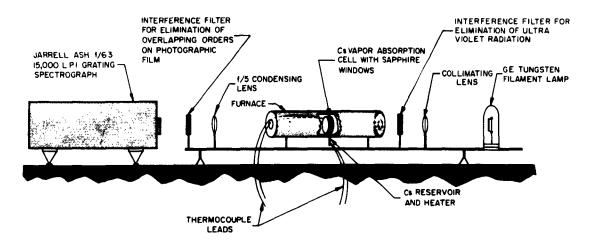


FIG. 3.9 SCHEMATIC ILLUSTRATION OF OPTICAL SYSTEM USED IN CESIUM ABSORPTION STUDIES

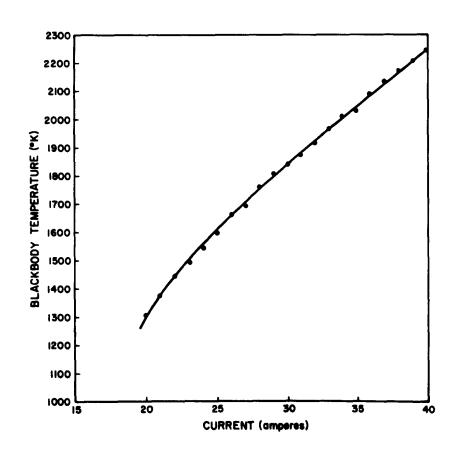


FIG. 3.10 TUNGSTEN FILAMENT TEMPERATURE CALIBRATION

The data presented in these figures give an excellent indication of the extent of increase of the absorption coefficient with vapor pressure.

These measurements are still in progress.

It is interesting to note the rapid increase of molecular absorption at the higher pressures. It has been shown (Ref. 21) by assuming a second virial equation of state for the cesium vapor that the ratio of the neutral cesium molecule density to atomic density is between  $10^{-3}$  to  $10^{-2}$ . Therefore, if the absorption cross section of the molecule is of the same order of magnitude as the atomic absorption cross section, the sudden increase in molecular absorption could be explained by a mechanism involving the production of cesium molecules in the vapor. The absorption at the resonance lines at 8521  $^{\rm A}$  and 8943  $^{\rm A}$  is producing excited cesium atoms which can combine to form the molecular ion.

$$Cs^* + Cs^* + KE \rightarrow Cs_2^+ + e$$

One of the possible complex ion loss mechanisms in the plasma will be recombination of the molecular ion with therma. lectrons. The product of this recombination may be a neutral molecule. This process could then have the effect of increasing the molecular to atomic ratio in the vapor. Loomis and Kusch (Ref. 11) have shown that absorption in the molecular band can also cause excitation of the molecule resulting in dissociation into an excited atom and a ground state atom. It is further possible that the excited cesium molecule produced by radiation absorption can produce ionic forms through complex collision mechanisms with other excited species existing in the plasma. Therefore, the molecular absorption may provide a further means of excitation and ultimately ionization in cesium vapor plasmas.

The rubidium impurity which is observed in Figure 3.6 represent an impurity of 0.1 percent and, therefore, through the cesium pressure range indicated on the figure, the rubidium constitutes a partial pressure range of  $6 \times 10^{-5}$  mm Hg to  $4 \times 10^{-3}$  mm Hg. Walter and Barratt (Ref. 20) indicate that for the pressure range of this work, there will be no overlap of the cesium molecular absorption bands with possible RbCs molecular absorption bands.

## 3.2 Resonance Decay Time Measurements

The experimental procedure is the same as that outlined by Alpert, McCoubrey, and Holstein (Ref. 22) for their measurements of resonance decay times in mercury vapor. The apparatus consists of a tungsten light source whose spectrum range is determined by a monochromator, an absorption cell, and a photo-multiplier. The light is turned on and off by a sectored disk rotating in front of the entrance slit of the monochromator. The radiation is then incident on the absorption cell and the diffuse resonance radiation coming from the absorption cell is indident on a photomultiplier. The resulting signal is amplified and displayed on an oscilloscope. The investigations in this area are still in progress.

## 3.3 <u>Ionization Processes in Cesium Vapor in the Presence of</u> Resonance Radiation

## 3.3.1 Theory

In a converter the resonance radiation is provided by the emitter where for gray body temperatures greater than approximately 1400°K. 8500 Å is well within the spectral range of the energy distribution. Figure 3.11 shows the total and spectral emissivity of several refractory materials which are commonly used as emitters. Notice that over the temperature range indicated, there can be almost an increase of two in the total emissivity. Indicating that as the temperature increases the radiating power will increase much more rapidly than its normal temperature dependence would allow. In order to calculate the percentage of atoms in the vapor which are excited to the resonance level by the incident radiation, the power available to be absorbed must first be determined.

The energy density  $(U_{\hat{D}})$  radiated by a black body emitter which is the energy available for absorption is:

$$U_v = \frac{8\pi h v^3}{c^3} \left[ \exp \frac{hv}{kT} - 1 \right]^{-1}$$
 (3.3.1)

in the wavelength region of interest https://kT is approximately 10. Therefore, the radiated energy density can be expressed as:

$$U_{v} = \frac{8\pi h v^{3}}{c^{3}} \exp\left(-\frac{hv}{kT}\right)$$
 (3.3.2)

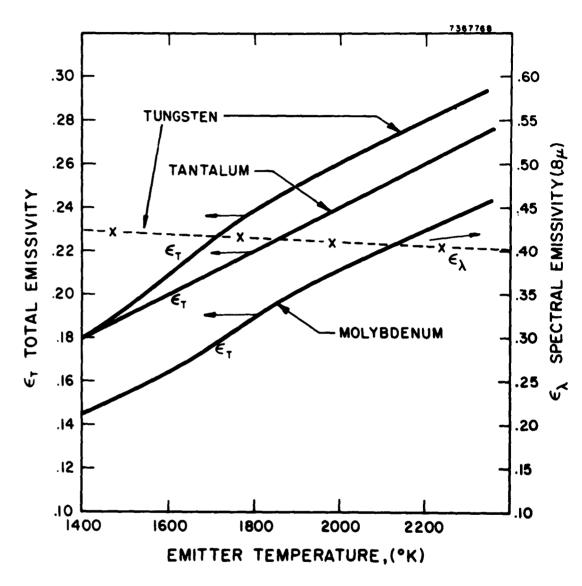


FIG. 3.11 EMISSIVITY OF SOME REFRACTORIES

where

h = Planck's constant

v = radiation frequency

c = speed of light

k = Boltzmann's constant

T = emitter temperature in degrees K

The power radiated  $(P_v)$  is,  $P_v = \frac{c}{4} \int_0^{\infty} U_v dv \qquad (3.3.3)$ 

Since the resonance absorption bandwidth,  $\delta v = v_2 - v_1$  used in the integral evaluation is for complete absorption, the power radiated in this bandwidth is the power absorbed in this bandwidth, therefore,

$$P_{abs} = \frac{2\pi h\epsilon}{c^2} \int_{v_1}^{v_2} v^3 e^{-\frac{hv}{kT}} dv \qquad (3.3.4)$$

where  $\epsilon$  = spectral emissivity of emitter near the resonance wavelength. To evaluate equation (3.3.4), Simpson's rule was used since 80 is small compared to  $v_0 = \frac{v_2 + v_1}{2}$ 

Therefore, the equation to determine power absorbed as a function of emitter temperature is:

$$P_{abs} = \frac{2\pi \epsilon h v_o^3}{C^2} (v_2 - v_1) e^{-hv_0}$$
 (3.3.5)

the results are given in Figure 3.12. The percentage excitation is obtained by considering an energy balance mechanism within the plasma. It will be assumed that the power absorbed in the plasma is equal to the power reradiated by the plasma.

$$P_{abs} = P_{rad}$$

$$P_{rad} = \frac{W_{rad}}{\tau}$$

where W = the energy radiated into the plasma and  $\tau$  = the radiation rad capture time or in other words the time in which the radiation is held in the plasma by successive absorption and decay before it reaches the side walls of the container. If we consider only resonance radiation from one level, say 8521 Å, then W is equal to the number of atoms in the excited state multiplied by the excitation energy. Therefore,

$$N = \frac{P_{abs}^{\tau}}{E_{ex}}$$
 atoms excited in a volume of 1 cm<sup>2</sup> xd gap (3.3.6)

For the results shown in Figure 3.12 and 3.13 a value of  $10^{-3}$  second for the capture decay time and 1 mm Hg cesium vapor pressure were used.

Alpert, McCoubrey and Holstein (Ref. 22) have done extensive work on imprisonment of resonance radiation in mercury vapor. They experimentally measured a decay time of resonance radiation in the vapor of 10<sup>-4</sup> second for a particle density of 10<sup>16</sup> atoms/cc. This value was lower than their theory predicted due to quenching effects by mercury isotopes. Since cesium is monoisotopic and can be obtained relatively pure, resonance decay times of 10<sup>-3</sup> second at pressures of 1 to 5 mm Hg are realistic. In order to determine the effect of shorter decay times on percentage excitation, Figure 3.14 was prepared. The percentage excitation in Figure 3.14 was determined by using the half height bandwidths of only the absorption band at 8521 A shown in Figure 3.5 From the figure the bandwidth for a cesium pressure of 1 mm Hg and 4 mm Hg is 80 Å and 175 Å respectively. The bandwidth for a cesium pressure of 10 mm Hg was found by extrapolation of the data from Figure 3.5 shown in Figure 3.15. All calculations were made assuming constant pxd relations (cesium vapor pressure times the emitter-collector distance). As Figure 3.14 indicates, a decrease in capture time can be counteracted by an increase in cesium vapor pressure to achieve a significant percentage of excitation.

It was indicated earlier that the maintenance voltage of low voltage, hot cathode arcs in the alkali vapors decreased as filament

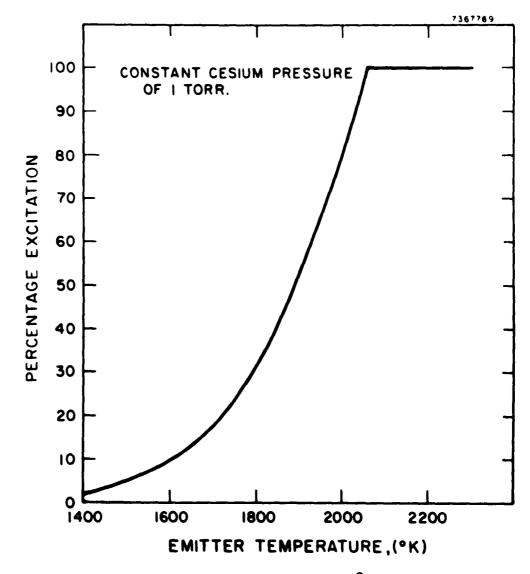


FIG. 3.13 EXCITED POPULATION AT 8521 & RESONANCE LEVEL IN CESIUM VAPOR DUE TO RADIATION FROM TUNGSTEN

EMITTER TEMP	CESIUM PRESSURE	8 λ	POWER AVAILABLE FOR ABSORPTION	PERCENTAGE EXCITATION	
				T=10 <sup>-3</sup> sec.	T=10 <sup>-4</sup> sec.
2200°K	.35 mm Hg	55 Å	.124 watts/cm <sup>2</sup>	100%*	12% ~
5500.K	l mm Hg	80 Å	.131 watts/cm <sup>2</sup>	100%*	15%
2200°K	4mmHg	175 Å	.282 watts/cm <sup>2</sup>	100%*	33%
5500.k	IOmmHg	300 Å	.487 watts/cm <sup>2</sup>	100 % <sup>#</sup>	57.2%

<sup>\*</sup> POWER AVAILABLE FOR EXCITATION EXCEEDS THAT NECESSARY FOR 100% EXCITATION IF 100% EXCITATION WERE POSSIBLE.

FIG. 3.14 PERCENTAGE EXCITATION AT 8521 Å AS A FUNCTION OF CESIUM PRESSURE AND CAPTURE TIME

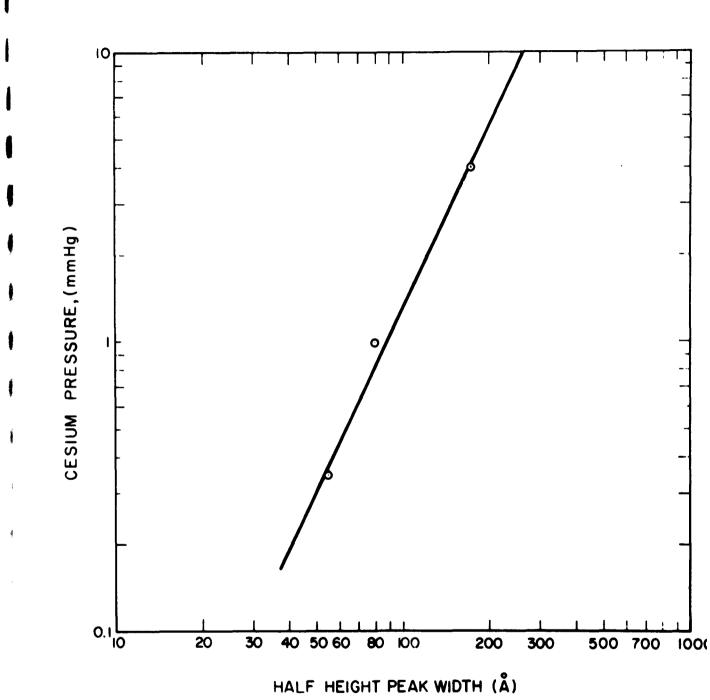


FIG. 3.15 HALF HEIGHT PEAK WIDTHS OF 8521 A RESONANCE LINE AS A FUNCTION OF CESIUM PRESSURE

temperature increased. This appears to be in excellent correlation with the increase in excited state population shown in Figure 3.13.

When the possibility of increasing the electrical conductivity of a gas by means of photoionization is considered, it is generally concluded that the rate at which positive ions are formed through the process of photon absorption by an atomic gas in an interelectrode space is bound to be insignificant when compared to the rates of other ionization processes, such as electron-atom impact. The principal difficulties usually cited are: (1) the production of high intensity radiation in the wavelength region which corresponds to the ionization continuum of the gaseous atoms, and (2) the weak absorption coefficient of an atomic vapor beyond the ionization limit.

These two limitations do seem to rule out the possibility of exploiting the photionization phenomenon as a means of neutralizing a negative space-charge, but only if it is assumed that photionization occurs exclusively through a one-step process. That is, the difficulties mentioned above are serious if the assumption is made that the only means by which visible or UV electromagnetic radiation can cause ionization is by directly exciting an atom from its ground state to a state lying in the continuum of levels above the ionization threshold. However, there exists strong evidence that this assumption is not valid or necessary when the ionization processes in certain metallic vapors are considered.

Houtermans (Ref. 23) and others have shown that ions can be produced in mercury vapor by illumination with 2537 Å radiation. The fact that the observed rate of ion formation was directly proportional to the square of the light intensity strongly suggested that ionization occurred through the cooperative action of two quanta of energy. It is interesting to note, however, that the energy sum of two quanta of 2537 Å radiation (2 x 4.9 e.v.) is less than the ionization energy of the mercury atom (10.4 e.v.). These observations of Houtermans bear a striking similarity to the results reported by Freudenberg on the ionization of cesium vapor upon the absorption of resonance radiation as indicated in Section 2. Weissler

(Ref. 24) has reported similar two-step ionization processes in cadmium and zinc vapors.

The relationship between atomic, molecular, and ionic term levels can best be illustrated by means of a Franck-Condon diagram as in Figure 3.16. For an excited Cs<sub>2</sub> molecule formed from a collision of two atoms in 2P states there exists a large density of states, consisting of a superposition of electronic and vibrational states. These states are characterized by a large variety of angular momentum and quantum mechanical symmetry properties, so that the requirements of energy coincidence and selection rules can be assumed to be satisfied for a significant number of these states.

In Figure 3.16 the total energy of a system composed of two cesium atoms in various states of excitation is plotted against the interatomic separation, r. The zero of energy is taken as the energy of two ground state atoms at rest with respect to one another and separated by an infinite distance. The solid and dashed lines represent the potential energy of the systems as the two atoms approach one another. The minima in the curves correspond to the fact that there is a net attraction for two cesium atoms at distances in the vicinity of the equilibrium bond distance of the diatomic molecule Cs<sub>2</sub>. The fact that repulsive states may also result when two atoms approach one another is indicated by the dashed lines.

The energy level diagram is simplified in the sense that only two of the many possible resulting states of the excited \*\*\* molecule,  $\operatorname{Cs}_2$ , are included. If it is assumed that one or the other of these two states is of the proper symmetry to be perturbed by the ground state of the molecular ion, then in order for ionization to occur it is only necessary to require that the potential energy curves of the two interacting systems come into close proximity or cross one another as the pairs of curves A and B, and B and C have been shown to do.

It should be emphasized that the details of the potential energy curves are not known and Figure 3.16 is intended to

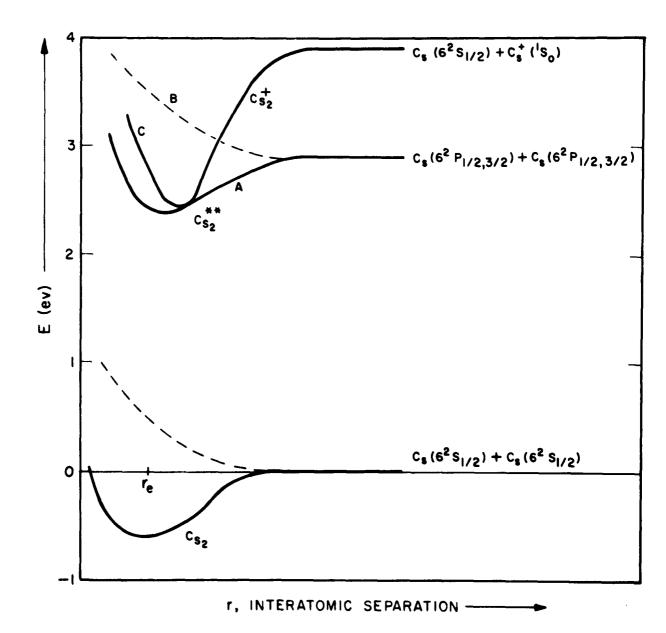


FIG. 3.16 POTENTIAL ENERGY CURVES OF THE CESIUM MOLECULE AND MOLECULAR ION

indicate the general features of the relationships that must exist between terms of the molecule and of the molecular ion in order that ionization (and by inference also two-step ionization) may take place.

## 3.3.2 Experimental Method

The experimental apparatus for ionization measurements is very similar to that for the absorption measurements shown in figure 3.8 except there is no need for the grating spectrograph. The ionization cell is very similar to the absorption cell except for a closer spacing between windows and the insertion of an electrically insulated tantalum mesh emitter and collector in front of the windows.

In operation the spacing between the meshes will be about .020" so that saturation thermionic emission can be drawn from the emitter at very low values of voltage. The graph of Figure 3.17 indicates the magnitude of applied voltage which is required to draw saturation emission for various mesh spacings. Since we expect saturation emission densities up to 10<sup>-4</sup> and 10<sup>-3</sup> amperes per square centimeter, the applied voltage necessary to eliminate space charge in the cell should be between 0.2 and 1.0 volts. Once saturation current is being drawn, the potential distribution between the meshes should be approximately linear and the external applied voltage should then be the voltage appearing between the vacuum levels of the emitter and collector meshes. This approach should allow a close determination of E/p in volts per centimeter-mm Hg in the region between the meshes. The measurement of the growth of current in the cell as a function of applied voltage then will lead to the determination of the first Townsend ionization coefficient in ion pairs per centimeter-mm Hg as a function of E/p. The measurements will be made over a range of cesium pressures both with and without injected resonance radiation in order to determine the role which resonance radiation plays in the ionization of cesium vapor.

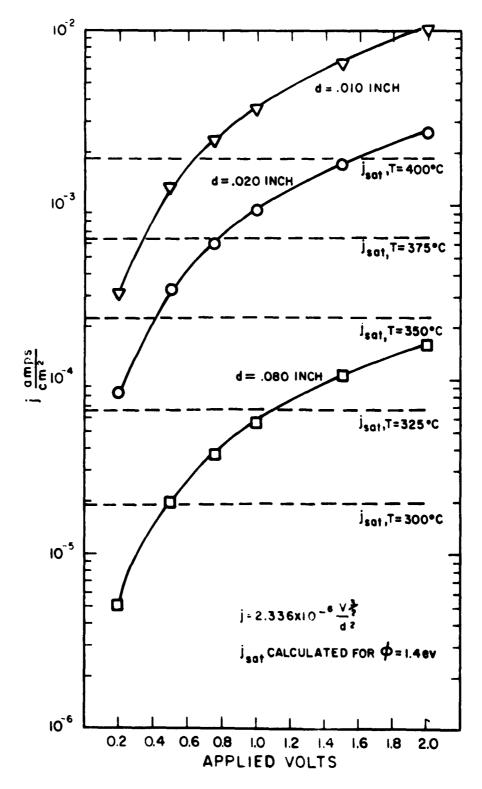


FIG. 3.17 SPACE CHARGE LIMITED CURRENT IN A PLANE PARALLEL DIODE, ELECTRODE SPACING AS A PARAMETER 3-28

#### 4. APPENDIX

### 4.1 Absorption Cell Fabrication

The absorption cell in essence consists of a cylindrical 304L stainless steel body I inch in diameter by I inch long with a cesium reservoir appendage extending from the side. The ends of the cylinder are sealed by sapphire windows which are brazed to the cylinder (see Figure 4.1).

The sapphire windows are first brazed to Kovar flanges which are then inert arc-welded to the cylinder. After the first two windows developed leaks, a series of test brazes were made in an effort to define time, temperature, and material requirements for successful window brazing. The photomicrographs in Figures 4.2, 4.3, 4.4, and 4.5 show in cross section, a series of nickel-titanium active alloy brazes between 97.5 percent Al<sub>2</sub>O<sub>3</sub> rings. The brazing time and temperature for the various samples are shown in Figure 4.6. It was felt that the high alumina rings would present approximately the same expansion and sealing problems as the sapphire windows, and would be much less expensive to experiment with. The titanium braze shim was a high purity type A-75 of 2.0 mil thickness. The nickel braze shim was a high purity type No. 499 (Driver Harris) of 0.5 mil thickness. This particular shim combination corresponds in the melt to a nickel-titanium alloy containing approximately 33 percent by weight of nickel. The melting point of the alloy is about 1015°C (see Figure 4.7).

The photomicrographs reveal two points of interest:

- The brazing time and temperature were too long and too high.
- 2. The nickel rich side of the 955°C eutectic point in the titanium-nickel phase diagram produces an alloy which is brittle and tends to be mushy throughout the braze cycle.

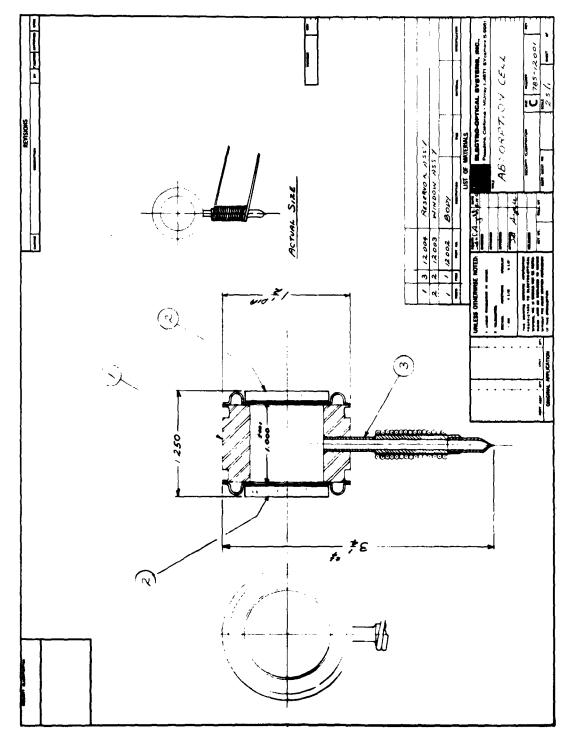
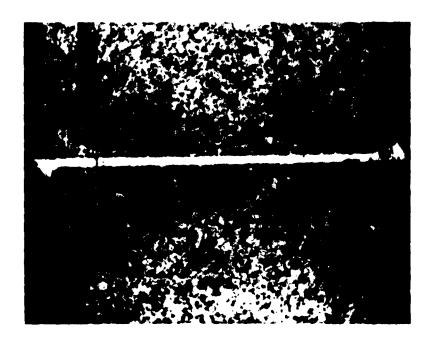
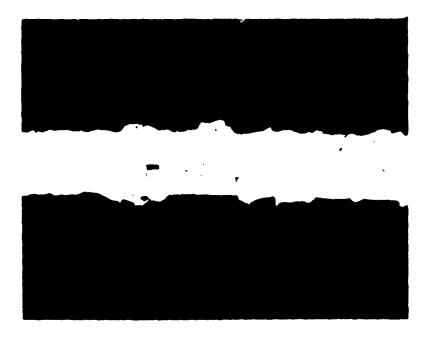


FIG. 4.1 ABSORPTION CELL

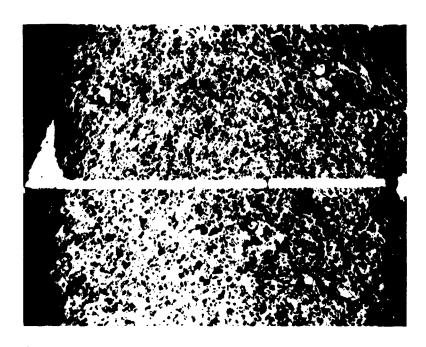


Sample No. 1 (48x)

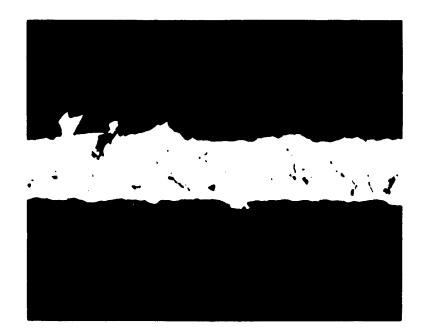


Sample No. 1 (384x)

FIG. 4.2 NICKEL-TITANIUM ACTIVE ALLOY BRAZE A1203 TO A1203 BRAZE TEMPERATURE 1050°C

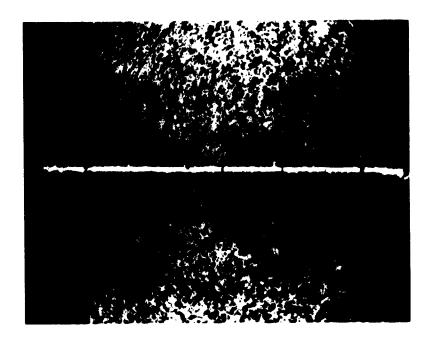


Sample No. 2 (50x)



Sample No. 2 (384x)

FIG. 4.3 NICKEL-TITANIUM ACTIVE ALLOY BRAZE A1203 TO A1203 BRAZE TEMPERATURE 1100°C

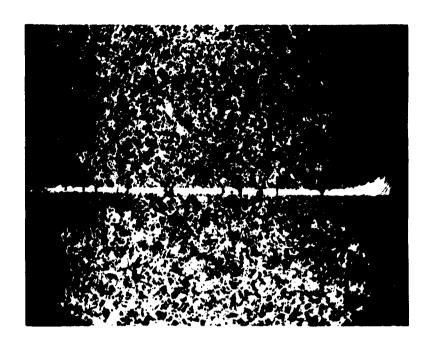


Sample No. 3 (48x)



Sample No. 3 (384x)

FIG. 4.4 NICKEL-TITANIUM ACTIVE ALLOY BRAZE Al<sub>2</sub>0<sub>3</sub> TO Al<sub>2</sub>0<sub>3</sub>
BRAZE TEMPERATURE 1185°C



Sample No. 4 (48x)



Sample No. 4 (384x)

FIG. 4.5 NICKEL-TITANIUM ACTIVE ALLOY BRAZE A1<sub>2</sub>0<sub>3</sub> TO A1<sub>2</sub>0<sub>3</sub>

BRAZE TEMPERATURE 1205°C

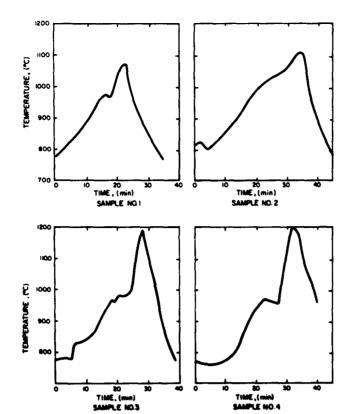


FIG. 4.6
NICKEL-TITANIUM ACTIVE ALLOY
BRAZE (A1<sub>2</sub>O<sub>3</sub>) TIME-TEMPERATURE
CURVES

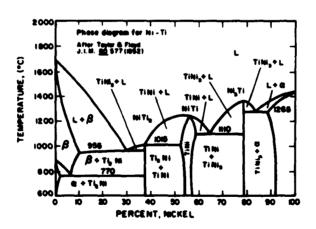


FIG. 4.7 NICKEL-TITANIUM

The conclusions to be drawn from these photomicrographs are:

- 1. Brazing temperatures should be kept between 950 and 1050°C.
- 2. Brazing time at temperature should be on the order of close to several minutes.
- 3. The appropriate braze shim thickness should be close to 5 to 1 or greater. That is, 5 parts titanium to 1 part nickel by volume. This puts the braze mi.ture at the 955°C nickel-titanium eutectic point (28.5 percent by weight nickel) or on the titanium rich side of this eutectic point.

An exploded view of the absorption cell and air oven used for temperature control is shown in Figure 4.8.

The spectral transmittance of the sapphire windows was determined in the wavelength range 0.25 - 2.5 microns. The measurements were made at normal incidence on a Perkin-Elmer Model 13-11 spectrophotometer. Typical of the results obtained is that depicted on a reduced scale in Figure 4.9.

It will be noted that the ultra-violet end of the transmission spectrum may be affected by the brazing operation if post-braze cleaning measures are not used. The degree of transmission reduction will be dependent upon the type of brazing material, the length of the brazing cycle, and the brazing temperature. Therefore, each absorption cell is calibrated for transmission in a detailed fashion before cesium is added.

Both the absorption and ionization cell were baked-out for 24 hours at temperatures of  $300^{\circ}$ C in a vacuum in which the pressure never exceeded 3 x  $10^{-6}$  mm Hg. After bake-out the cells were loaded with vacuum distilled cesium in the same apparatus in which the bake-out took place.

## 4.2 Ionization Chamber Fabrication

As shown in Figure 4.10, the conization measurement chamber consists essentially of two insulated close spaced tantalum meshes surrounded by a cesium environment. The sapphire window: allow the intro-

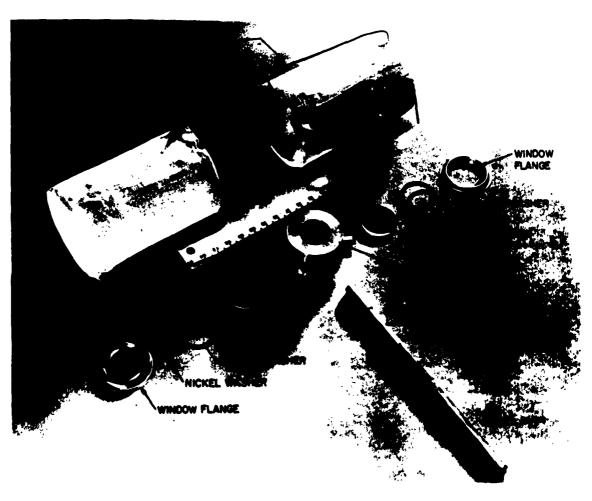


FIG. 4.8 EXPLODED VIEW OF EOS ABSORPTION CELL

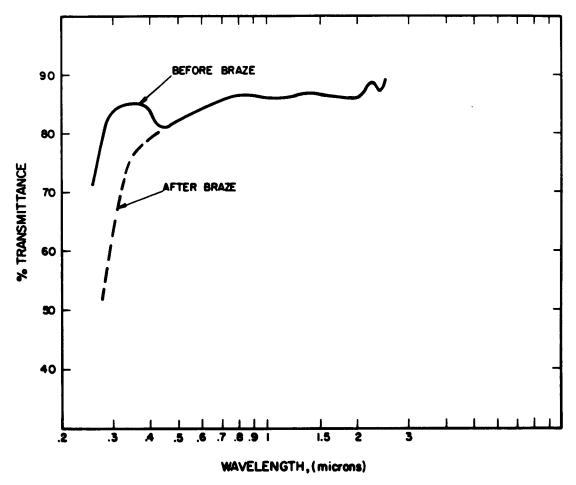
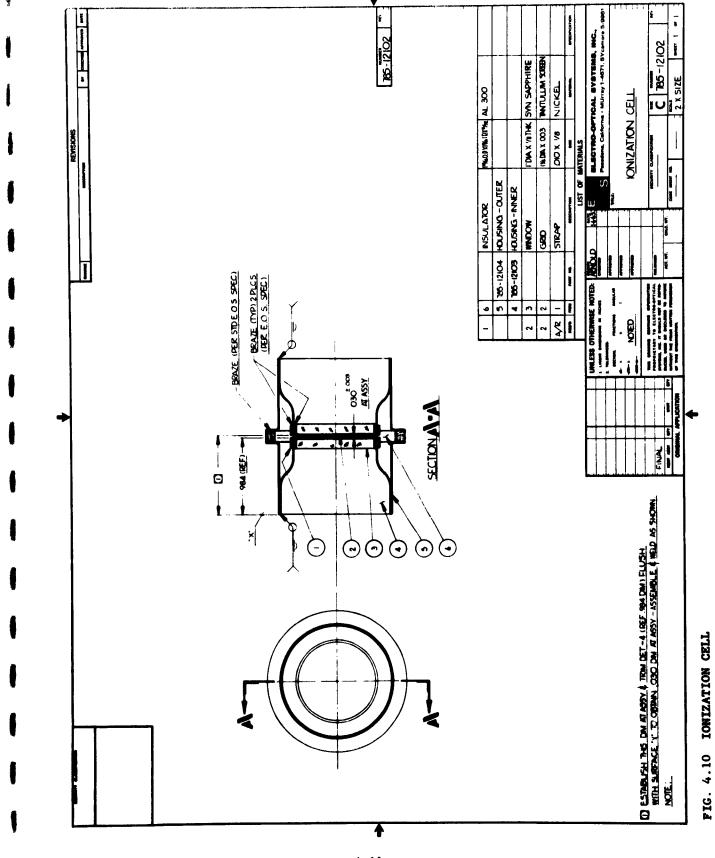


FIG. 4.9 SPECTRAL TRANSMITTANCE CHARACTERISTIC OF A 3mm THICK SAPPHIRE WINDOW



4-11

duction of resonance radiation for cesium vapor into the region between the meshes. The meshes serve as identical work function emitter and collector. This latter property is due to the fact that the tantalum meshes are operated at the same temperature and cesium arrival rate. The chamber body is constructed from Kovar and the sapphire windows are brazed to Kovar flanges as described in Section 4.1.

## 4.3 Experimental Data Tabulation

Typical values of the absorption coefficients that have been obtained for the broadened 8521 A cesium absorption line are included in the following tables and Figure 4-11. The absorption properties of a cesium vapor at a temperature, T, can be determined by the equation:

$$I_{0} = I_{0} e^{-k_{\lambda}(T) L}$$
 (4.3.1)

where L is the length of the absorbing vapor. For example, at  $318^{\circ}$ C the value of  $k_{\lambda}$  is given as 1.25 cm<sup>-1</sup> for the wavelength 8579 Å. Therefore, the attenuation of a beam of radiation at this wavelength passing through a cell length of 8 mm amounts to a factor of about 1/e, since,

$$I = I_0 e^{-1.256}$$
 (.8)  $2 I_0 e^{-1}$ 

The absorption coefficients were obtained from photographic film which was calibrated with the aid of a logarithmic sector disc, a 5000 RPM motor, a uniform temperature tungsten ribbon lamp and a microdensitometer. These values of the absorption coefficients are subject to verification by further measurements and the studies are being extended to higher temperatures and shorter path length absorption cells.

TABLE I

CESIUM TEMP. - 194°C: CELL ENVIRONMENT TEMP. 395°C

<u>\lambda</u>	k <sub>\(\lambda\)</sub> (cm <sup>-1</sup> )
8653.6	.0298
8618.0	0.1066
8600.1	0.1340
8582.3	0.1882
8564.5	0.3440
8546.7	0.755
8528.9	Total absorption at a path length of 2.54 cm.
8511.0	Total absorption at a path length of 2.54 cm.
8493.2	0.690
8475.4	0.381
8457.6	0.260
8439.8	0.206
8421.9	0.184
8404.12	0.131
8386.3	0.131
8172.5	0.077
7994.3	0

TABLE II

CESIUM TEMP. - 238°C: CELL ENVIRONMENT TEMP. 395°C

<u>x (Å)</u>	$\frac{k_{\lambda}(cm^{-1})}{}$
8699.6	0.166
8610.5	0.232
8592.7	0.279
8574.9	0.413
8557.1	0.729
8539.2	1.960
8521.4	Total absorption at a path length of 2.54 cm.
8503.6	Total abs rotion at a path length of 2.54 cm.
8485.8	1.000
8468.0	0.648
8450.1	0.440
8432.3	0.368
8414.5	0.295
8343.2	0.208
8254.1	0.166
8165.0	0.084
8075.9	0
7986.8	0

TABLE III

CESIUM TEMP. - 277°C: CELL ENVIRONMENT TEMP. 395°C

<u>\(\hat{A}\)</u>	$\frac{k_{\lambda}(cm^{-1})}{}$
8707.2	0.270
8672.0	0.270
8636.3	0.351
8618.5	0.417
8600.7	0.531
8582.8	0.902
8565.0	2.030
8529.4	Total absorption at a path length of 2.54 cm
8493.8	Total absorption at a path length of 2.54 cm
8476.0	0.986
8458.2	0.777
8440.4	0.637
8422.5	0.529
8404.7	0.513
8386.9	0.434
8351.3	0.351
8297.8	0.306
8262.2	0.253
8173.1	0.158
8084.1	0

TABLE IV

CESIUM TEMP. - 318°C: CELL ENVIRONMENT TEMP. 395°C

<u>λ (Å)</u>	$\frac{k_{\lambda}(cm^{-1})}{k_{\lambda}(cm^{-1})}$
8632.4	0.482
8614.5	0.630
8596.17	0.849
8578.9	1.256
8561.1	Total absorption at a path length of 2.54 cm
8525.4	Total absorption at a path length of 2.54 cm
8489.8	Total absorption at a path length of 2.54 cm
8472.0	Total absorption at a path length of 2.54 cm
8454.2	0.999
8436.3	0.818
8418.5	0.700
8400.7	0,592
8382.9	0.538
8365.1	0.484
8347.2	0.364
8311.6	0.333
8169.0	0.315
8080.9	0
7991.8	0

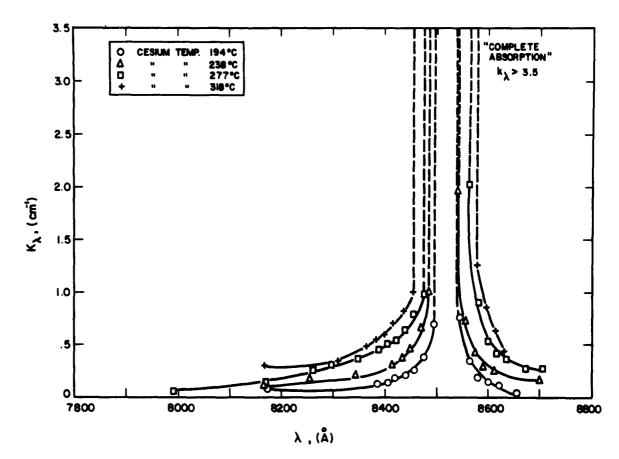


FIG. 4.11 ABSORPTION COEFFICIENT AS A FUNCTION OF WAVELENGTH

## 5. PRINCIPAL CONTRIBUTORS

A. O. Jensen, H. R. Moore, D. H. Pollock, and J. A. Duardo were principal contributors to this program during the past work period.

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Electro-Optical Systems, Inc., Pasadena, Calif. A. O. Jensen Contract Nonr-3805(00) Thermionic converters Ionization processes Radiation absorption Cestum vapor plasma 1.11 1264 tribute significantly to the energy input to a cesium vapor plasma. At high cesium pressure (greater than 0.3 mm Hg) and emitter temperatures (greater than 1800 k) the absorption of resonance radiation may be the most significant energy input into a cesium vapor thermionic converter plasma leading to the formation of the cesium molecular ion as the dominant ionic species in the plasma according to the relationship: posed that appears to be applicable to the cestum pressure range and other conditions found in cestum vapor thermionic converters. It has been found that the atomic resonance absorption bandwidths at the \$321 Å and \$943 Å resonance lines are much greater than previously reported. A molecular absorption band near 7600 Å was also observed which can con-The results of an investigation into the absorption of resonance radiation in cesium vapor and the effect of resonance radiation on the lonization effects as in cesium vapor plasmas are reported. A model for the dominant ionization processes is pro-Measurements on resonance radiation decay times and cesium ionization in the presence of resonance radiation are still in progress. (over) (heat of formation Cs<sub>2</sub>)-⊕Cs<sub>2</sub> + e Navy, Office of Naval Research, Department of the Nashington 25, D. C. Rpt. No. 3130 Annual. DOCKINST IONIZATION PROCESS IN CESTUM VAPOR TREPRIONIC CONVERTER FLASMAS, Annual report. June 63, 61 p. incl illus, tables, 24 refs. \*5) + 5) \*5) I. Contract Nonr-3803(00)
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